

X-ray magnetic scattering study of three-dimensional magnetic order in the quasi-one-dimensional antiferromagnet $\text{Nd}_2\text{BaNiO}_5$

A. Zheludev and J. P. Hill

Department of Physics, Brookhaven National Laboratory, Upton, New York 11973-5000

D. J. Buttrey

University of Delaware, Newark, Delaware 19716

(Received 3 April 1996)

We report a resonant x-ray-scattering determination of order parameter of the Nd sublattice in the quasi-one-dimensional antiferromagnet, $\text{Nd}_2\text{BaNiO}_5$. We find that long-range order develops below $T_N=48$ K, and that the staggered magnetization continues to grow down to $T=10$ K. This is in contrast with the behavior of the Ni sublattice, which quickly saturates, as determined previously by neutron-scattering. We find this behavior is modeled well by a self-consistent mean-field theory. Large resonant enhancements were obtained at both the Nd L_{II} and L_{III} absorption edges, with the largest count rates being achieved at the L_{II} edge, where structure consistent with magnetic diffraction anomalous fine structure was observed. A double peak observed at the L_{III} resonance is also discussed. [S0163-1829(96)06234-0]

I. INTRODUCTION

The study of one-dimensional (1D) magnets has occupied a central place in the field of magnetism over the years because of the importance of quantum fluctuations in such systems and the tractability of certain of the simplest models. To date, the majority of the work performed has been investigations of quasi-1D systems in which every effort is made to approach the 1D limit, and there has been less work on the crossover to 3D behavior as the coupling between the 1D elements of the systems is increased. Recently however, a new class of transition-metal oxides has been synthesized which allows the introduction of magnetic or nonmagnetic ions between 1D chains of Ni spins. These ions mediate an interchain interaction and hence a systematic variation of this coupling is possible through chemical substitution, and the 1D–3D crossover may be studied in some detail. As a result they have attracted a great deal of attention. The members of the series are the isostructural nickelates with the general formula $L_2\text{BaNiO}_5$, where $L=Y$ or a magnetic rare earth, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, and Tm.^{1,2} In this work, we report an x-ray magnetic scattering study of one member of this series, $\text{Nd}_2\text{BaNiO}_5$.

The $L_2\text{BaNiO}_5$ compounds feature antiferromagnetic $S=1$ Ni^{2+} chains with spinless ($L=Y$) or magnetic ($L=\text{rare earth}$) ions in between, which makes them model quasi-one-dimensional (1D) mixed-spin antiferromagnets. They have an orthorhombic structure (space group $Immm$), with cell constants $a \approx 3.8$ Å, $b \approx 5.9$ Å, and $c \approx 11.7$ Å, as first described by Müller-Bushbaum.³ Strongly distorted NiO_6 octahedra share apical oxygens to form linear chains along the a crystallographic axis. The in-chain antiferromagnetic exchange constant, as determined by inelastic neutron scattering, is relatively large, $|J| \approx 250$ K.^{4,5} For nonmagnetic L ($L=Y$) the interchain interactions are negligible and Y_2BaNiO_5 is a near-ideal example of a quantum-disordered linear-chain $S=1$ antiferromagnet exhibiting a spin-singlet

ground state and a Haldane gap in the magnetic excitation spectrum.^{6,7,4} Further, it does not display any long-range magnetic order, at least down to 1.2 K. Substituting magnetic rare earth ions for Y increases the interchain coupling and all the $L \neq Y$ members of the series order antiferromagnetically with the 3D Néel temperatures T_N ranging from 24 to 80 K (see Ref. 8, and references therein).

Intriguingly, recent inelastic neutron-scattering studies of $\text{Pr}_2\text{BaNiO}_5$ (Ref. 5) and $\text{Nd}_2\text{BaNiO}_5$ (Ref. 9) have shown that Haldane-like gap excitations are present not only in Y_2BaNiO_5 , but also in those $L_2\text{BaNiO}_5$ members that undergo a transition to a 3D ordered state. Thus these materials exhibit behavior intermediate between true 1D and true 3D phenomena. The onset of long-range antiferromagnetic order does not, as might be expected, eliminate 1D gap excitations propagating on the Ni sites, but in fact leads to an increase in the gap-gap energy. These phenomena are not yet fully understood. A better understanding of the underlying physics of these unusual systems will require a detailed knowledge of the magnetic structure and the temperature dependence of the order parameter of both the Ni and rare earth sites.

In this work, we report a resonant magnetic x-ray scattering study of $\text{Nd}_2\text{BaNiO}_5$. As discussed below, this technique is element specific and one can measure *directly* the temperature dependence of the ordered moment of the Nd^{3+} independently of any Ni contribution. This is in contrast to neutron-scattering measurements for which a trial magnetic structure must be calculated and the ordered moments of the two sublattices are then extracted from intensities of a number of Bragg peaks. In this sense, the work reported here is complementary to the neutron-scattering studies of Sachan *et al.*⁸ In addition, x-ray scattering offers the advantages that only small samples are required; that high resolution is obtained as a natural consequence of the intrinsic collimation of the synchrotron x-ray source; and that the small cross section ensures that even for strong magnetic Bragg peaks, the intensity is not extinction limited and a reliable measurement

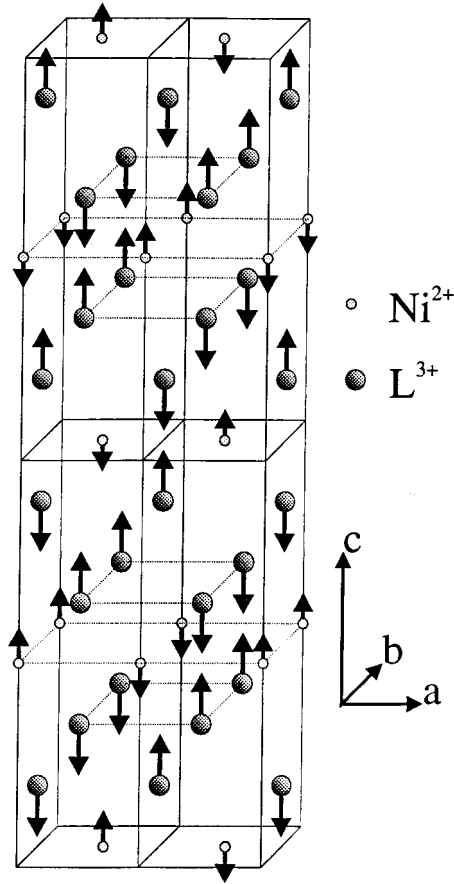


FIG. 1. The ordered spin structure of $L_2\text{BaNiO}_5$ as deduced from neutron-scattering results of Ref. 8. In the present work, $L=\text{Nd}$. The Ba and O atoms are omitted for clarity.

of the order parameter is possible. The sample studied here was of high crystallographic quality and large count rates were obtained, allowing a precise determination of the temperature dependence of the Nd ordered moment.

We find that the temperature dependence of the sublattice magnetizations in $\text{Nd}_2\text{BaNiO}_5$ can be adequately described by a simple self-consistent mean-field model, with the Ni moment quickly saturating below T_N and the Nd moment continuing to grow down to the lowest temperatures. We discuss these results in relation to the temperature dependence of the gap in the Ni-chain modes. Large resonant enhancements were observed at both the Nd L_{II} and L_{III} absorption edges and furthermore, the energy dependence of the magnetic scattering suggests a possible hybridization of the Nd $4f$ - $5d$ orbitals. The magnetic Bragg peaks were found to be resolution limited and we conclude that the Néel state does indeed have long-range order.

The Néel state in $L_2\text{BaNiO}_5$ has been investigated by neutron diffraction for $L=\text{Nd}$, Ho, Er, and Tm.^{10,11,2,8} In particular, for $\text{Nd}_2\text{BaNiO}_5$ Sachan *et al.* have shown that the magnetic structure is close to collinear with the ordered spins pointing along the c crystallographic axis.⁸ This structure is shown in Fig. 1. The magnetic unit cell is doubled relative to the crystallographic cell along both the a and c axes, and the onset of long-range magnetic order manifests itself in the appearance of new magnetic Bragg reflections of type

$((2n+1)/2 \ 0 \ (2m+1)/2) \ (n, m \text{ integer})$. Antiferromagnetic ordering occurs at $T_N=48$ K and the Ni moments were found to be essentially saturated below $T=30$ K. In contrast, the magnetic moment on Nd continues to increase down to $T=10$ K.⁸

Before presenting the results of this study, we first discuss x-ray magnetic scattering. Briefly, magnetic scattering by x rays may be considered in two limits. For incident photon energies far from an atomic absorption edge, there is weak spin-dependent scattering; smaller by a factor of order $(\hbar\omega/mc^2)^2$ than the Thomson (charge) scattered intensity.¹² This is nonresonant scattering and is typically weak even at a synchrotron source. However, by tuning the incident photon energy to an absorption edge, the resonant terms, arising from the second-order terms in the perturbation expansion of the cross section, may dominate. This scattering may be sensitive to the magnetic moment if the particular edge chosen involves the excitation of an electron to a magnetically ordered shell, or to an exchange split orbital.^{13,14} Under such conditions, coherent magnetic Bragg diffraction occurs and large count rates may be obtained. The scattered intensity is proportional to the square of the staggered magnetization in the case of an antiferromagnet, and correlation lengths may be measured in the usual way.^{15,16} To date, the largest enhancements in the hard x-ray region have been observed at the L edges of the rare earths and the M edges of the actinides. The technique is inherently element specific, since the resonant contributions of a given species tend to dominate the scattered signal. In the present case, we are able to measure the Nd ordered moment by tuning to the Nd L_{II} and L_{III} edges, with little contribution from the Ni moment expected.

II. RESULTS

The experiments were carried out at the X22C beamline at the National Synchrotron Light Source. A toroidal focusing mirror and a double bounce Ge(111) monochromator were used to obtain a beam of size $\approx 1 \times 1 \text{ mm}^2$ at the sample position. Most of the measurements were performed with a Ge(111) flat analyzer crystal, which has the dual function of reducing the background signal and providing good longitudinal resolution. The overall energy resolution was ≈ 5 eV full width at half maximum and the longitudinal reciprocal-space resolution was 0.0027 \AA^{-1} . The sample studied was an irregularly shaped single crystal of $\text{Nd}_2\text{BaNiO}_5$ of dimensions $\approx 0.5 \times 3 \times 4 \text{ mm}^3$, taken from the same batch as the sample used by Sachan *et al.* in their neutron-diffraction work. The crystal mosaic was found to be of the order of 0.005° . The sample was mounted in a He-filled Be can on the end of the cold finger of a closed-cycle He refrigerator with a base temperature of $T=10$ K.

Resonant magnetic scattering was observed when the incident photon energy was tuned to the Nd L_{II} and L_{III} absorption edges, at 6722 and 6208 eV, respectively. The largest enhancement was obtained at the L_{II} edge. In Fig. 2(a) we plot the scattered intensity as a function of incident photon energy on tuning through the L_{II} edge. The data were taken at the $(0.5, 0, 8.5)$ antiferromagnetic Bragg peak at $T=10$ K, and are plotted on a linear scale in the inset and a semilogarithmic scale in the main panel. A count rate of 3000 cps at

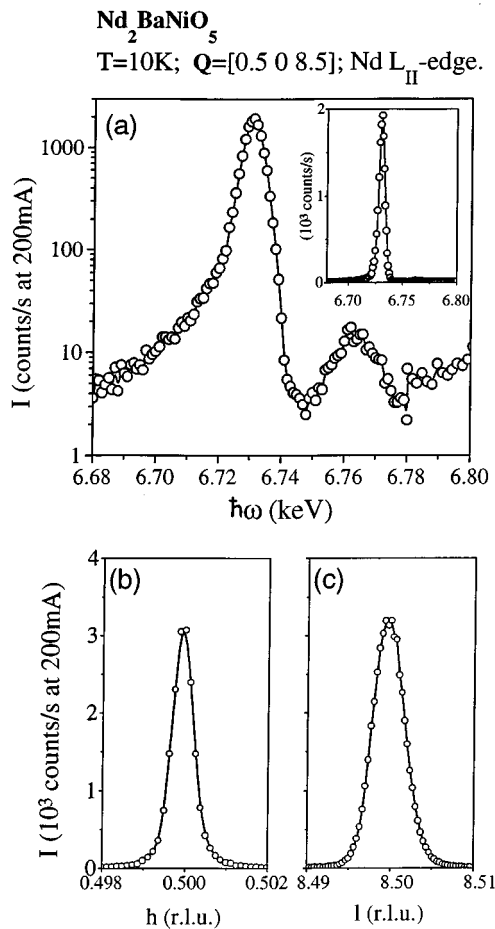


FIG. 2. (a) Energy dependence of the magnetic Bragg peak (0.5 0 8.5) as the incident photon energy is tuned through the L_{II} absorption edge. Inset: Same data on a linear scale. Data were taken at $T=10$ K. (b) (h 0 8.5) and (c) (0.5 0 l) scans through the magnetic Bragg peak, taken at the peak of the L_{II} resonance at $T=10$ K.

200 mA was achieved at the peak of the L_{II} resonance. Scans taken along the h and l directions through the antiferromagnetic Bragg peak are shown in Figs. 2(b) and 2(c), respectively. The widths are the same as that of equivalent scans through a charge Bragg peak and we therefore conclude that the magnetic Bragg peak is resolution limited and that the Néel state indeed has long-range order. We may set a lower bound on the correlation lengths from the half width of these scans: We obtain $\xi_c \geq 375$ Å and $\xi_a \geq 770$ Å.

The structure evident in the energy dependence of the Nd L_{II} resonance of Fig. 2 warrants further comment. The energy line shape displays a marked asymmetry with a tail on the low-energy side. In addition, there is some fine structure on the high-energy side of the resonance. It seems likely that the asymmetry is the result of the interference between the resonant and nonresonant magnetic scattered amplitudes. Such effects have been observed previously, for example in Ho.¹⁷ In Ho, it was found that the asymmetry increased as the scattering angle increased because of the angular dependence of the nonresonant amplitude.¹⁸ For the spin structure of Fig. 1, the angular dependence is expected to be stronger than in the case of Ho, which may explain why the asymmetry is so pronounced in Fig. 2(a).

The peak on the high-energy side of the spectra of Fig.

2(a) however cannot result from the interference of the non-resonant and resonant amplitudes, which can only produce a minimum in the scattered intensity. We speculate that this feature is in fact the first oscillation of the anomalous fine structure associated with the interference of the photoelectrons wave function with the surrounding atoms. Such fine structure is well known in absorption measurements, and has long been used as a probe of local order (x-ray-absorption fine structure, or XAFS). Recently this technique has been combined with diffraction by studying the fine structure observed in the Bragg scattered intensity as the incident photon energy is tuned through an absorption edge. This has been termed diffraction anomalous fine structure (DAFS).¹⁹ We believe that the oscillation in Fig. 2(a) represents magnetic DAFS. XAFS measurements provide information on the local structural order, for example the bond lengths, types, and disorder. Extending the technique to DAFS, Straiger *et al.*¹⁹ pointed out that as well as containing the same local information, DAFS has the additional benefit of providing spatial or site selectivity when different regions, or sites, in the sample contribute differently to Bragg peaks. The observation of magnetic DAFS reported above, allows the same conclusions to be drawn about magnetically inequivalent sites. In our view, the intensity gains (factors of 10 or more) available at insertion device beamlines, relative to the bending magnet source of X22C should make such experiments feasible.

Resonant magnetic scattering was also observed at the Nd L_{III} edge. The intensities of the (1 0 7) charge and (0.5 0 8.5) magnetic Bragg peaks measured at $T=10$ K in the vicinity of the L_{III} resonance are plotted against incident photon energy in Figs. 3(a) and 3(b), respectively. The intensity of the (1 0 7) charge peak shows a characteristic dip at the resonance, as a result of the interference of the anomalous scattering terms in the cross section. In contrast, the (0.5 0 8.5) magnetic peak is resonantly enhanced. The positions of the dip for charge scattering and the maximum for magnetic scattering coincide with a steep increase in the fluorescence from the sample Fig. 3(c). The resonant enhancement at the L_{III} edge is significantly smaller than that at the L_{II} edge. The ratio of the intensities, the branching ratio, is (without absorption corrections) $I_{L_{II}}/I_{L_{III}} \approx 50$. This is similar to the ratio obtained in previous work on Nd₂CuO₄.²⁰ Work on elemental Nd reported a branching ratio of 6 after correction for absorption.²¹ The relative strengths of the L_{II} and L_{III} resonances are not fully understood and in fact existing theories predict that they should be comparable in magnitude.²² However, it has been observed that there is a systematic trend amongst the rare earths in which the light rare earths (less than half-filling of the 4f shell) generally exhibit a larger L_{II} resonance and the heavy rare earths a larger L_{III} enhancement. Gd, with an exactly half-filled 4f shell fits this pattern with a branching ratio of around unity.²³ Some of these branching ratios were tabulated by Watson *et al.*²¹

A striking feature seen at the magnetic Bragg position in the vicinity of the Nd L_{III} edge is the double-peaked structure of the resonance [Fig. 3(b)]. This is in marked contrast to the L_{II} absorption edge [inset in Fig. 2(a)], where only a single peak is observed. The origin of the doublet in the L_{III} resonance is not clear. One hint perhaps, lies in the observation that the splitting between the doublet components, ≈ 5 eV, is

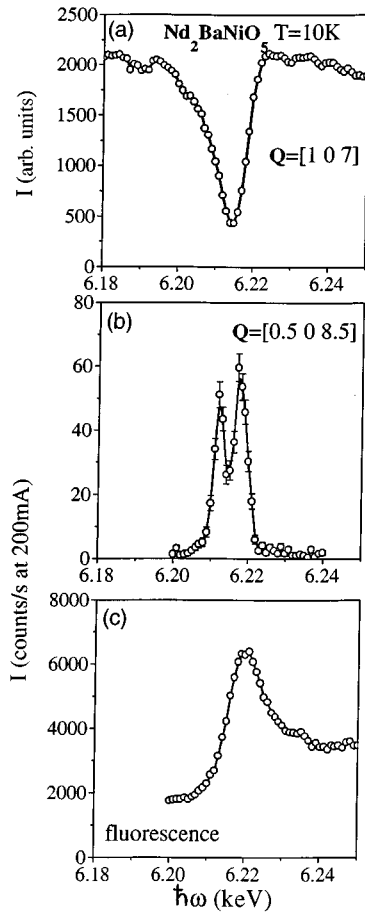


FIG. 3. Energy dependences near the L_{III} edge. (a) A charge Bragg peak (1 0 7). (b) A magnetic Bragg peak (0.5 0 8.5). (c) The fluorescence, as monitored by measuring the background intensity with no analyzer far from any Bragg peaks.

similar to the energy separation of $5d$ and $4f$ electrons in Nd.²⁴ One possibility then, is that at least one component may be attributed to quadrupolar terms in the cross section. While in the dipole approximation the L_{II} and L_{III} resonances couple $2p_{1/2}$ and $2p_{3/2}$ states to $5d$, respectively, for quadrupolar transitions the final states are $4f_{7/2}$ and $4f_{5/2}$. For the $2p_{3/2}$ initial state (L_{III} edge) transitions to both $4f_{7/2}$ and $4f_{5/2}$ are permitted by quadrupolar selection rules, but only the $2p_{1/2} \rightarrow 4f_{5/2}$ transition is allowed for the L_{II} resonance. The selection rules alone cannot fully explain the observed behavior, and we simply note that the quadrupolar selection rules are less strict for the L_{III} resonance than for L_{II} . This would require quadrupolar scattering to be roughly equal in magnitude to the dipolar and, while magnetic resonant quadrupolar scattering has been previously observed in holmium,^{13,17} the quadrupolar contribution was found to be significantly weaker than the dipolar term, as expected. In addition, no such strong quadrupolar resonances were observed in either Nd (Ref. 21) or Nd_2CuO_4 at the L_{III} edge. There is however an alternative explanation. The noncentrosymmetric structure of Nd_2BaNiO_5 allows for the possibility of mixing the $5d$ and $4f$ orbitals, which in turn permits dipole transitions to states with largely $4f$ character. Were such $4f$ - $5d$ hybridization to occur only with the $5d_{5/2}$ derived states and not the $5d_{3/2}$ states, then the dipole selection

rules would permit only transitions from the $2p_{3/2}$ level to the new level, i.e., they could only occur at the L_{III} edge. This would explain the lack of two peaks at the L_{II} edge and the relative strength of those at the L_{III} edge. In analogy with the situation in Ho, one might expect that the presence of the core hole would pull the largely $4f$ unoccupied level below the $5d$ band so that the lower energy feature would correspond to this transition. We emphasize that these ideas are speculative and further work, for example polarization analysis or theoretical calculations, is required to confirm or refute these ideas.

Having found large magnetic Bragg intensities at $T > T_N$, we hoped that resonant critical scattering would be observable at $T > T_N$. Unfortunately, it could not be detected in our experiments. This is one down-side of the high Q -resolution x-ray diffraction provides: diffuse features are often practically impossible to see. We have also carried out a search for nonresonant scattering, with the incident photon energy set to $E_i = 7.23$ keV. At $T = 10$ K, a weak peak of ≈ 1 cps above background was observed. This sets an upper limit to the contribution of the Ni moment to the scattered intensity in the data reported below. In comparison, at $T = 10$ K, at the peak of the Nd L_{II} resonance, a count rate of approximately 3000 cps was obtained. Thus for all practical purposes, the Ni contribution may be ignored.

Temperature-dependent measurements of the Nd^{3+} order parameter were carried out at the (0.5 0 8.5) antiferromagnetic reciprocal-space zone center, with the incident photon energy tuned to the Nd L_{II} absorption edge. At each temperature the peak was first centered in a (longitudinal) l scan [Fig. 2(b)] and the integrated intensity was measured in a rocking (transverse) h scan [Fig. 2(c)]. The (0.5 0 8.5) integrated intensity is plotted against temperature in the inset in Fig. 4. The main panel in Fig. 4 shows the temperature dependence of the (arbitrarily scaled) Nd^{3+} magnetic moment (open circles), which is proportional to the square root of the Bragg integrated intensity. The results obtained in neutron diffraction experiments⁸ are shown in solid symbols, circles for Nd^{3+} and squares for Ni^{2+} . While the Ni^{2+} moments rapidly reach saturation upon cooling, the Nd^{3+} order parameter continues to increase down to 10 K. Between 10 and 40 K this temperature dependence is almost linear.

III. DISCUSSION

García-Matres *et al.*²⁵ have analyzed their dc magnetic-susceptibility data for several L_2BaNiO_5 systems using a simple mean-field model. Following this analysis, we have found that a similar approach works surprisingly well in describing the temperature dependence of the Nd^{3+} ordered moment measured in our experiments. The low site symmetry for Nd^{3+} ($mm2$) splits the tenfold degenerate ground state into five Kramers doublets. Only the lowest-energy doublet is populated at low temperatures and Nd^{3+} may be treated as an effectively spin- $\frac{1}{2}$ ion with a certain gyromagnetic ratio g_{Nd} . Since the magnitude of the single-ion anisotropy on the Ni sites is a subject of some debate,²⁶ we have arbitrarily chosen to use the two-level approximation for the Ni^{2+} as well, introducing the effective gyromagnetic ratio g_{Ni} . We further assume that the magnetization of the Ni^{2+} and Nd^{3+} sublattices can be calculated using the Brillouin

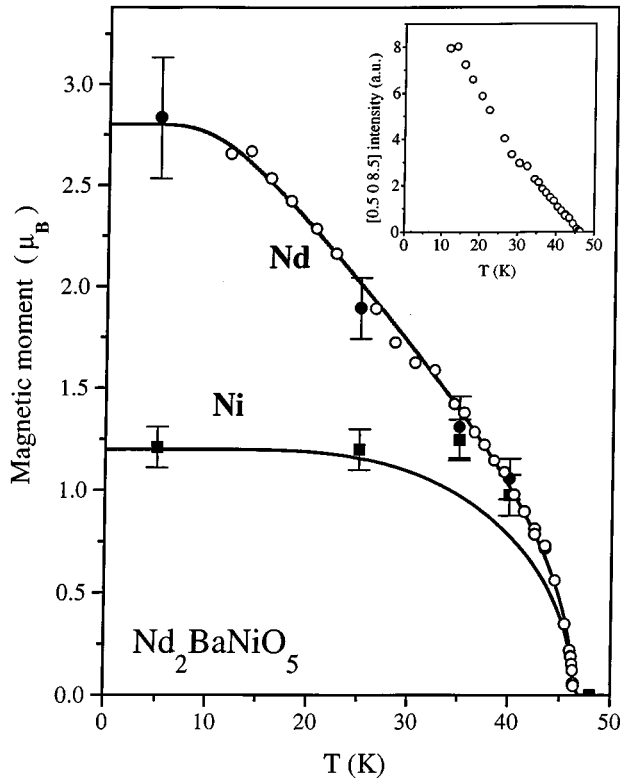


FIG. 4. Temperature dependence of the Nd ordered moment (open circles) as derived from measurements taken at the L_{II} resonance. The data have been arbitrarily scaled by an overall multiplicative factor to compare with the (sparser) neutron results of Sachan *et al.* (Ref. 8) (closed circles). The closed squares represent the Ni ordered moment as extracted from the neutron fits. The solid lines represent the moments obtained from a self-consistent mean-field theory as discussed in the text.

function for paramagnetic ions:

$$M_{\text{Ni}} = \frac{1}{2} g_{\text{Ni}} \mu_B \tanh \left[\frac{(1/2) g_{\text{Ni}} \mu_B H_{\text{Ni}}}{k_B T} \right],$$

$$M_{\text{Nd}} = \frac{1}{2} g_{\text{Nd}} \mu_B \tanh \left[\frac{(1/2) g_{\text{Nd}} \mu_B H_{\text{Nd}}}{k_B T} \right], \quad (1)$$

where μ_B is the Bohr magneton, k_B is Boltzmann's constant, and H_{Ni} and H_{Nd} are the effective self-consistent local fields induced by the surrounding magnetic ions on the Ni and Nd sites, respectively:

$$H_{\text{Ni}} = \alpha M_{\text{Ni}} + \beta M_{\text{Nd}},$$

$$H_{\text{Nd}} = \gamma M_{\text{Nd}} + \beta M_{\text{Ni}}. \quad (2)$$

Here α , β , and γ are, respectively, the effective Ni-Ni, Ni-Nd, and Nd-Nd coupling constants. To obtain the temperature dependences $M_{\text{Nd}}(T)$ and $M_{\text{Ni}}(T)$ Eqs. (1) and (2) are solved self-consistently at each temperature. Fixing the gyromagnetic ratios to the values deduced from neutron-diffraction experiments⁸ ($g_{\text{Nd}} = 5.6$ and $g_{\text{Ni}} = 2.4$), and assuming the Nd-Nd coupling to be negligible²⁵ (i.e., $\gamma = 0$), we require only three adjustable parameters to fit the experimental temperature dependence of the Nd^{3+} moment: two

coupling constants α and β and an overall scaling factor. A least-squares refinement results in a very good fit, reproducing all the main features of the experimental curve. The temperature dependences of the Ni^{2+} and Nd^{3+} ordered moments calculated from the refined parameter values are shown in solid lines in Fig. 4. The refined values for α and β are $2.97(3) \times 10^5 \text{ Oe}/\mu_B$ and $3.72(3) \times 10^4 \text{ Oe}/\mu_B$. Due to the phenomenological nature of this mean-field approach, the absolute values of these parameters are not very meaningful, although their relative magnitudes demonstrate the dominance of Ni-Ni interactions. As suggested by García-Matres *et al.*,²⁵ the Ni^{2+} moments become saturated upon cooling much faster than those of Nd^{3+} . Below $T = 35 \text{ K}$ the magnetization of the Nd^{3+} sublattice may be viewed as being induced by a temperature-independent staggered field generated by the ordered Ni^{2+} spins. This produces a broad linear region in $M_{\text{Nd}}(T)$. A similar induced ordering mechanism for rare earth moments has been previously observed in other systems, such as Nd_2CuO_4 and Pr_2CuO_4 .^{27,28}

It is important to emphasize that the mean-field approach described above is greatly oversimplified. *A priori*, there is no reason for the mean-field approximation to be applicable to a strongly 1D system like $\text{Nd}_2\text{BaNiO}_5$. For example, in our treatment we have assumed the Nd-Nd interactions are negligible. In fact, these exchange pathways are believed to be essential for completing a 3D spin network and enabling the spontaneous magnetic ordering.⁵ However, it may be that a mean-field treatment is adequate for the Nd^{3+} moments due to the dominance of Ni-Ni coupling and the rapid ordering of the Ni^{2+} sublattice. This may explain why such a good agreement with experiment is obtained. For the Ni^{2+} sites on the other hand, the experimentally observed approach to saturation (Fig. 4, solid squares)⁸ is visibly more rapid than predicted by the mean-field model (Fig. 4, lower solid line), demonstrating the limitations of this approach in describing the Ni ordering.

As far as critical behavior is concerned, our measurements fail to detect any deviations of the critical exponent β from the Ginsburg-Landau value $\beta = 0.5$. For real 3D magnets β is expected to be reduced ($\beta \approx 0.35$ for the 3D Heisenberg model) and even more so for lower-dimensionality systems. Obviously, in our temperature-dependent measurements the proximity of T_N could not be covered with sufficient precision, and the measured “ $\beta = 0.5$ ” refers to the temperature dependence outside the critical region.

The temperature dependence of the magnetic moment on the Ni^{2+} and L^{3+} sites is central to understanding the temperature dependence of the energy gap in the Haldane-like Ni-chain magnetic excitation branches in $\text{Nd}_2\text{BaNiO}_5$ (Ref. 9) and $\text{Pr}_2\text{BaNiO}_5$ (Ref. 5). In these references it was shown that the gap mode reflects spin fluctuations on the Ni sites, with no contribution from L^{3+} . Above T_N the energy gap is $\approx 11 \text{ meV}$ and shows little temperature dependence. Below the Néel temperature, the gap increases linearly with decreasing T . This behavior apparently continues well below the temperature at which the Ni^{2+} moments become saturated. At these temperatures the only changes in the static magnetization arise from the L^{3+} sites. As the present work demonstrates, the L^{3+} ordered moment steadily increases, at least down to $T_N/5$. Therefore, one possible explanation for

the temperature dependence in the Ni-chain modes is that they are influenced by an effective temperature dependent staggered field induced by the progressive ordering of the L^{3+} spins. This scenario needs further investigation. In particular, theoretical models of such interesting mixed-spin quasi-1D quantum antiferromagnets as the $L_2\text{BaNiO}_5$ family would greatly facilitate the analysis of these experiments.

IV. SUMMARY

Resonant magnetic x-ray scattering was observed in $\text{Nd}_2\text{BaNiO}_5$ at both the L_{III} and L_{II} absorption edges. The temperature dependence of the Nd^{3+} order parameter was measured and found to grow continually down to the lowest temperature studied, in contrast the Ni moment which is believed to saturate quickly. The temperature dependence of the ordered moments of the two sublattices was modeled successfully with a simple mean-field theory. Magnetic DAFS “wiggles” in the resonance spectrum were clearly

seen, suggesting the possibility of future work at more intense x-ray sources. Resonant magnetic scattering is an accurate and *element-specific* technique which may provide a powerful complement to neutron experiments, in particular for two sublattice systems such as this one. In application to $L_2\text{BaNiO}_5$ materials, x rays may be the only probe suitable for investigating the rich field-temperature phase diagram of $\text{Gd}_2\text{BaNiO}_5$,^{25,29} due to the extremely strong neutron absorption of Gd.

ACKNOWLEDGMENTS

The authors would like to thank M. Blume and J. M. Tranquada for illuminating discussions and K. Kakurai for sharing some of his unpublished results on Y_2BaNiO_5 . Work at Brookhaven National Laboratory was supported by the Division of Material Sciences, U.S. Department of Energy, under Contract No. DE-AC02-76CH00016.

- ¹D. J. Buttrey, J. D. Sullivan, and A. L. Rheingold, *J. Solid State Chem.* **88**, 291 (1990).
- ²E. García-Matres *et al.*, *J. Solid State Chem.* **103**, 322 (1993).
- ³S. Schiffer and H. Müller-Buschbaum, *Z. Allg. Chem.* **10**, 523 (1986).
- ⁴T. Yokoo, T. Sakaguchi, K. Kakurai, and J. Akimitsu, *J. Phys. Soc. Jpn.* **64**, 3651 (1995).
- ⁵A. Zheludev, J. M. Tranquada, T. Vogt, and D. J. Buttrey, *Phys. Rev. B* **54**, 6437 (1996); *Europhys. Lett.* (to be published).
- ⁶J. Darriet and L. P. Regnault, *Solid State Commun.* **86**, 409 (1993).
- ⁷J. F. DiTusa *et al.*, *Physica B* **194-196**, 181 (1994).
- ⁸V. Sachan, D. J. Buttrey, J. M. Tranquada, and G. Shirane, *Phys. Rev. B* **49**, 9658 (1994).
- ⁹A. Zheludev, J. M. Tranquada, T. Vogt, and D. J. Buttrey, preceding paper, *Phys. Rev. B* **54**, 7210 (1996).
- ¹⁰J. A. Alonso *et al.*, *Solid State Commun.* **76**, 467 (1990).
- ¹¹A. Salinas-Sánchez, R. Sáez-Puche, J. Rodríguez-Carvajal, and J. L. Martínez, *Mater. Chem. Phys.* **31**, 145 (1992).
- ¹²M. Blume, *J. Appl. Phys.* **57**, 3615 (1985).
- ¹³D. Gibbs, D. R. Harshman, E. D. Isaacs, D. B. McWhan, D. Mills, and C. Vettier, *Phys. Rev. Lett.* **61**, 1241 (1988).
- ¹⁴J. P. Hannon, G. T. Trammel, M. Blume, and D. Gibbs, *Phys. Rev. Lett.* **61**, 1245 (1988).
- ¹⁵J. P. Hill and D. F. McMorrow, *Acta Crystallogr. A* **52**, 236 (1996).
- ¹⁶J. Luo, Ph.D. thesis, Rice University, 1994.
- ¹⁷D. Gibbs *et al.*, *Phys. Rev. B* **43**, 5663 (1991).
- ¹⁸M. Blume and D. Gibbs, *Phys. Rev. B* **37**, 1779 (1988).
- ¹⁹H. Straiger, J. O. Cross, J. J. Rehr, L. B. Sorensen, C. E. Bouldin, and J. C. Woicik, *Phys. Rev. Lett.* **69**, 3064 (1992).
- ²⁰J. P. Hill, A. Vigliante, D. Gibbs, J. L. Peng, and R. L. Greene, *Phys. Rev. B* **52**, 6575 (1995).
- ²¹D. Watson, E. M. Forgan, D. Fort, W. J. Nuttall, and W. G. Stirling, *Phys. Rev. B* **53**, 726 (1996).
- ²²M. D. Hamrick, Ph.D. thesis, Rice University, 1994.
- ²³C. Detlefs, A. I. Goldman, C. Stasis, P. C. Canfield, B. K. Cho, J. P. Hill, and D. Dabbs, *Phys. Rev. B* **53**, 6355 (1996).
- ²⁴J. K. Lang, Y. Baer, and P. A. Cox, *J. Phys. F* **11**, 121 (1981).
- ²⁵E. García-Matres, J. L. García-Muñoz, J. L. Martínez, and J. Rodríguez-Carvajal, *J. Magn. Magn. Mater.* **149**, 363 (1995).
- ²⁶The energy splitting of the Haldane triplet in Y_2BaNiO_5 determined by inelastic neutron-scattering on powder samples by Darriet and Regnault (Ref. 6) suggests that the anisotropy is of a strong easy-plane character. More recent single-crystal studies by the authors of Ref. 4 seem to indicate that only a very weak easy-plane component is present [K. Kakurai (private communication)].
- ²⁷M. Matsuda *et al.*, *Phys. Rev. B* **42**, 10 098 (1990).
- ²⁸M. Matsuda *et al.*, *Phys. Rev. B* **45**, 12 548 (1992).
- ²⁹M. N. Popova, I. V. Paukov, Yu. A. Hadjiiskii, and B. V. Mill, *Phys. Lett. A* **203**, 412 (1995).